

PL/2-22807

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF

Group Art Unit: 1793

PAOLO BALLIELLO

Examiner: SHUANGYI ABU ALI

SERIAL NO. 10/537,020

FILED: June 1, 2005

FOR: DUST-FREE PIGMENT COMPOSITIONS
OBTAINABLE BY SPRAY-DRYING

Commissioner for Patents

Washington, D.C. 20231

DECLARATION UNDER 37 CFR 1.132

I, Paolo Balliello, a citizen of Italy, residing in I-55044 Marina di Pietrasanta, Italy, hereby declare:

That the University of Padua, Italy, awarded me the degree of Doctor of Sciences (chemistry) in 1969;

That for 7 years I have been employed as a plant chemist in chemical production within the companies Hoechst and Ciba-Geigy;

That for 28 years I have then been in charge of many chemical development projects within Ciba-Geigy and Ciba Specialty Chemicals Inc. at Maastricht, Netherland and Basle, Switzerland;

That I have 35 years of experience in the field of organic dyestuffs and pigments,

especially in the development and scaling up of methods for the production of organic pigments and pigment preparates, as well as new pigment preparates;

That I retired from Ciba Specialty Chemicals Inc. on August 31, 2007.

That I am the inventor or co-inventor of 11 patents, including US Patents in fields closely related to that of the above-identified application;

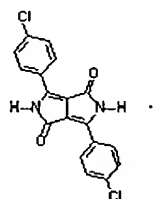
That I have read carefully the instant application 10/537,020 as well as GB-842,791 (Ambler / ICI), GB-1,176,217 (Zwahlen & Steinlin / Ciba), US patents 3,728,143 (Pollard), 3,843,380 (Beyn), 4,264,552 (McMahon & Newton), 5,082,498 (Kurtz et al.) and 5,681,836 (Schneider et al.), and that I am very familiar with the subject matter thereof;

That the experiments described in the following have been carried out by me or under my supervision and the evaluation of the test results has been done by myself.

EXPERIMENTAL

Comparative experiments were done in close analogy to the instant examples, using

Colour Index Pigment Red 254:



Instant sample A according to the invention was prepared by using as the binder, based on the total pigment composition weight, 4.5% by weight of Armeen[®] T (tallow alkyl amine) and 0.5% by weight of Metolose[®] SM-100 (methyl cellulose with approximately 30 % methoxy content).

Comparative sample B was prepared according to the prior art Beyn (US-3,843,380 / column 5 / lines 45-48).

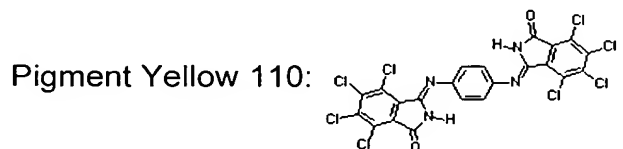
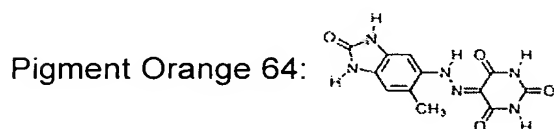
Comparative sample C was prepared according to a purely hypothetical combination of prior art Beyn (US-3,843,380) with example 7 of Ambler (GB-842,791).

Sample	Methocel® 65HG	Trioctylamine	Armeen® T	Metolose® SM100	dispersibility in PVC
A			4.5%	0.5%	good
B	5%				poor (‡)
C	5%	2%			very poor (‡)

(‡) More than 5% increase in color strength after additional, cold 2-rolls extrusion, measured on white reductions (0.2% color pigment + 5% TiO₂). After normal hot processing, comparative sample B was 11% and comparative sample C was 17% below the values obtainable after full dispersion.

The pigment according to the invention (sample A) was non-dusting while nevertheless having a good dispersibility in PVC. Though the total amount of binder (including trioctylamine) is the same in sample B as in sample A, the dispersibility of comparative sample B comprising only modified cellulose was poor. Comparative sample C, obtained according to a purely hypothetical combination of prior art, which is however not taught in the prior art, had the worst, much poorer dispersibility.

Furthermore, I also performed comparative experiments in low density polyethylene (LDPE), using organic pigments of other structural classes:



These two pigments were each treated in close analogy to sample A, B and C, however with the amounts of binders indicated in the tables below.

According to the standard test procedure used at Ciba, samples D, E, F, G and H were tested by rolling 0.5 parts of organic pigment (including the binders), 5.0 parts of titanium white and 94.5 parts of LDPE, and the chroma, dispersibility and colour strength of the LDPE test plates obtained after standard processing were measured. The results were as follows (as compared with the standard quality, binder-free dry pigment used for preparing samples D, E, F, G and H):

C. I. Pigment Orange 64

Sample	Methocel® 65HG	Trioctyl- amine	Armeen® T	Metolose ® SM100	chroma	color strength	dispersibility (visually)
D					53.2	100%	standard
E			4.5%	0.5%	61.9	180%	better
F	5%				18.3	6%	unsatis- factory
G	5%	2%			23.5	11%	unaccep- table
H	2.5%	1%			48.0	70%	bad

C. I. Pigment Yellow 110

Sample	Methocel® 65HG	Trioctyl- amine	Armeen® T	Metolose ® SM100	chroma	color strength	dispersibility (visually)
J					51.5	100%	standard
K			4.5%	0.5%	52.5	105%	equal
L	5%				33.6	28%	unaccep- table
M	5%	2%			31.4	23%	unaccep- table
N	2.5%	1%			47.9	78%	equal

The instant samples E and K comprising a binary mixture of a long chain alkyl amine (Armeen® T) and a cellulose derivative (Metolose® SM100) clearly led to much superior results in LDPE. To the contrary of the binary binder mixture according to the invention, a negative effect of trioctylamine was observed in combination with Methocel® 65HG (the binder according to Beyn US-3,843,380), as illustrated by the chroma, color strength and partly dispersibility of samples G, H, M and N.

DISCUSSION OF RESULTS

These results show clearly that modified cellulose thickeners as taught by Beyn (US-3,843,380) are inadequate to improve the dispersibility of pigments, and that further addition of trioctylamine as taught by Ambler (GB-842,792) does not improve, but on the contrary worsen the dispersibility when combined with the modified cellulose thickeners of Beyn (US-3,843,380).

According to my invention, it is critical to combine the modified cellulose with a suitably chosen second binder: the combination of a specific modified cellulose with

a second binder of formulae $Q-N \begin{smallmatrix} R_3 \\ R_4 \end{smallmatrix}$, $Q-\overset{\overset{O}{\parallel}}{C}-N \begin{smallmatrix} R_3 \\ R_4 \end{smallmatrix}$ or $Q-\overset{\overset{O}{\parallel}}{C}-O-R_3$ leads to a huge

synergistic improvement, while the combination with trioctylamine leads to a decrease in dispersibility. Such unexpected synergism between Metolose[®] SM100 and a fatty alkyl amine (Armeen[®] T) is demonstrated by above experimental data. Please further consider that the quantity of the specific modified cellulose may instantly be neither less than 5% nor more than 60% of the total binder, while there is

from 40 to 95% of the compound of formulae $Q-N \begin{smallmatrix} R_3 \\ R_4 \end{smallmatrix}$, $Q-\overset{\overset{O}{\parallel}}{C}-N \begin{smallmatrix} R_3 \\ R_4 \end{smallmatrix}$ or $Q-\overset{\overset{O}{\parallel}}{C}-O-R_3$.

CONCLUSIONS

Hence, the prior art documents GB-842,791 (Ambler / ICI), GB-1,176,217 (Zwahlen & Steinlin / Ciba), US patents 3,728,143 (Pollard), 3,843,380 (Beyn), 4,264,552 (McMahon & Newton), 5,082,498 (Kurtz et al.) and 5,681,836 (Schneider et al.), either alone or even in combination with each other, fail to disclose or suggest my invention, which is therefore clearly novel and not obvious.

I further declare that all statements made herein of my own knowledge are true and

that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

Signed, in Pittsboro, this 25th day of August, 2008

P. Balliello

Paolo Balliello